

Amendments to the Specification:

Please amend the specification as follows:

On page 20, between lines 22 and 23, please add the following twenty-two new paragraphs:

Two suitable non-metallic multi-layer optical films described in the above-mentioned PCT Publication No. WO 01/96104 are (a) a birefringent dielectric multilayer film that reflects at least 50% of light in a band at least 100 nm wide in a wavelength region of interest, wherein the film is heat set at a temperature sufficient to render the film capable of shrinking to conform without substantial wrinkling to a substrate having a compound curvature and (b) birefringent dielectric multilayer film that reflects at least 50% of light in a band at least 100 nm wide in a wavelength region of interest, wherein the film is heat set at a temperature sufficient to enable the film to shrink at least about 0.4 % in both in-plane directions upon heating.

To reflect over a wide band, the various layers in the birefringent dielectric multilayer films preferably have varying relative thicknesses, referred to herein as a layer thickness gradient, which are selected to achieve the desired bandwidth of reflection. In one embodiment, the layer thickness gradient may be linear, in which the thickness of the layer pairs increases at a constant rate across the thickness of the films, so that each layer pair is a certain percent thicker than the thickness of the previous layer pair. In another embodiment, the layer thickness may decrease, then increase, then decrease again from one major surface of the films to the other, or may have an alternate layer thickness distribution designed to increase the sharpness of one or both bandedges, as described in U.S. Patent No. 6,157,490.

In another embodiment, the birefringent dielectric multilayer films include an extended bandedge two component IR reflecting film construction having a six layer alternating repeating unit as described in U.S. Patent No. 5,360,659. This construction suppresses the unwanted second, third, and fourth order reflections in the visible wavelength region of between about 380 to about 700 nm, while reflecting light in the infrared wavelength region of between about 700 to about 2000 nm. Reflections higher than fourth order will generally be in the ultraviolet, not visible, region of the spectrum or will be of such a low intensity as to be unobjectionable. The film comprises alternating layers of first (A) and second (B) polymeric materials in which the six layer alternating repeat unit has relative optical thicknesses of about

.778A.111B.111A.778B.111A.111B. The use of only six layers in the repeat unit results in more efficient use of material and is relatively easy to manufacture. In this construction, it is again desirable to introduce a repeat unit thickness gradient across the thickness of the film.

In yet another embodiment, the two component birefringent dielectric multilayer films may comprise a first portion of alternating layers comprising the six layer alternating layer repeating unit which reflects infrared light of wave lengths between about 1200 - 2000 nm and a second portion of alternating layers having an AB repeat unit and substantially equal optical thicknesses which reflect infrared light of wavelengths between about 700 - 1200 nm. Such a combination of alternating layers is called a hybrid design and results in reflection of light across the infrared wavelength region. The layer thicknesses of both portions of alternating layers can be adjusted to place the reflecting band within the infrared spectrum so as to minimize any perceived color change with angle.

The birefringent dielectric multilayer optical films may also include more than two distinguishable polymers. A third or subsequent polymer might be fruitfully employed as an adhesion-promoting layer between the first polymer and the second polymer within an optical stack, as an additional component in a stack for optical purposes, as a protective boundary layer between optical stacks, as a skin layer, as a functional coating, or for any other purpose. As such, the composition of a third or subsequent polymer, if any, is not limited.

In an alternate embodiment, birefringent dielectric multilayer films including more than two distinguishable polymers can be made. This extends the reflecting band further into the IR to increase the amount of IR reflection while minimizing color in the visible region of the spectrum, which results from reflection due to higher order harmonics. Examples of such films include those described in U.S. Patent No. RE 34,605. The RE '305 patent describes a multilayer optical interference film including three diverse substantially transparent polymeric materials, A, B, and C and having a repeating unit of ABCB. The layers have an optical thickness of between about 90 nm to about 450 nm, and each of the polymeric materials has a different index of refraction, n_i .

To produce a birefringent dielectric multilayer film that reflects a broad bandwidth of wavelengths in the solar infrared range (e.g., reflection at from about 700 to about 2000 nm), a layer thickness gradient is introduced across the thickness of the film, and the layer thicknesses

preferably increase monotonically across the thickness of the film. Preferably, for a three component system, the first polymeric material (A) differs in refractive index from the second polymeric material (B) by at least about 0.03, the second polymeric material (B) differs in refractive index from the third polymeric material (C) by at least about 0.03, and the refractive index of the second polymeric material (B) is intermediate the respective refractive indices of the first (A) and third (C) polymeric materials. Any or all of the polymeric materials may be synthesized to have the desired index of refraction by utilizing a copolymer or miscible blend of polymers.

Another useful film design is described in U.S. Patent No. 6,027,260. Optical films and other optical bodies are described which exhibit a first order reflection band for at least one polarization of electromagnetic radiation in a first region of the spectrum while suppressing at least the second, and preferably also at least the third, higher order harmonics of the first reflection band, while the percent reflection of the first order harmonic remains essentially constant, or increases, as a function of angle of incidence.

This is accomplished by forming at least a portion of the optical body out of polymeric materials A, B, and C which are arranged in a repeating sequence ABC, wherein A has refractive indices n_x^A , n_y^A , and n_z^A along mutually orthogonal axes x, y, and z, respectively, B has refractive indices n_x^B , n_y^B , and n_z^B along axes x, y and z, respectively, and C has refractive indices n_x^C , n_y^C and n_z^C along axes x, y, and z, respectively, where axis z is orthogonal to the plane of the film or optical body, wherein $n_x^A > n_x^B > n_x^C$ or $n_y^A > n_y^B > n_y^C$, and wherein $n_z^C \geq n_z^B \geq n_z^A$. Preferably, at least one of the differences $n_z^A - n_z^B$ and $n_z^B - n_z^C$ is less than or equal to about -0.05.

By designing the film or optical body within these constraints, at least some combination of second, third and fourth higher-order reflections can be suppressed without a substantial decrease of the first harmonic reflection with angle of incidence, particularly when the first order reflection band is in the infrared region of the spectrum.

In an alternate embodiment, the birefringent dielectric multilayer films may comprise a first portion of alternating layers comprising a multicomponent optical design that reflects infrared light of wavelengths between about 1200-2000 nm while minimizing higher order reflections that contribute to visible color, and a second portion of alternating layers having an

AB repeat unit and substantially equal optical thicknesses which reflect infrared light of wavelengths between about 700-1200 nm. This hybrid design may be provided as described, for example, in U.S. Patent No. 5,360,659, but has broader application in that it is useful with any of the multicomponent optical designs described herein. The layer thicknesses of both portions of alternating layers can be adjusted to place the reflecting band within the infrared spectrum to minimize any perceived color change with angle.

The first and second optical layers and the optional non-optical layers of the birefringent dielectric multilayer films are typically composed of polymers such as, for example, polyesters. The term “polymer” will be understood to include homopolymers and copolymers, as well as polymers or copolymers that may be formed in a miscible blend, for example, by coextrusion or by reaction, including, for example, transesterification. In general, the use of comonomers should not substantially impair the stress optical coefficient or retention of birefringence after stretching. In practice, these restrictions impose an upper limit on the comonomer content, the exact value of which will vary with the choice of comonomer(s) employed. Some compromise in optical properties may be accepted, however, if comonomer incorporation results in improvement of other properties. The terms “polymer”, “copolymer”, and “copolyester” include both random and block copolymers.

Polyesters for use in the multilayer reflective mirrors and polarizers of the present invention generally include carboxylate and glycol subunits and are generated by reactions of carboxylate monomer molecules with glycol monomer molecules. Each carboxylate monomer molecule has two or more carboxylic acid or ester functional groups and each glycol monomer molecule has two or more hydroxy functional groups. The carboxylate monomer molecules may all be the same or there may be two or more different types of molecules. The same applies to the glycol monomer molecules. Also included within the term “polyester” are polycarbonates derived from the reaction of glycol monomer molecules with esters of carbonic acid.

Suitable carboxylate monomer molecules for use in forming the carboxylate subunits of the polyester layers include, for example, 2,6-naphthalene dicarboxylic acid and isomers thereof; terephthalic acid; isophthalic acid; phthalic acid; azelaic acid; adipic acid; sebacic acid; norbornene dicarboxylic acid; bi-cyclooctane dicarboxylic acid; 1,6-cyclohexane dicarboxylic acid and isomers thereof; t-butyl isophthalic acid, trimellitic acid, sodium sulfonated isophthalic

acid; 2,2'-biphenyl dicarboxylic acid and isomers thereof; and lower alkyl esters of these acids, such as methyl or ethyl esters. The term "lower alkyl" refers, in this context, to C1-C10 straight-chained or branched alkyl groups.

Suitable glycol monomer molecules for use in forming glycol subunits of the polyester layers include ethylene glycol; propylene glycol; 1,4-butanediol and isomers thereof; 1,6-hexanediol; neopentyl glycol; polyethylene glycol; diethylene glycol; tricyclodecanediol; 1,4-cyclohexanedimethanol and isomers thereof; norbornanediol; bicyclo-octanediol; trimethylol propane; pentaerythritol; 1,4-benzenedimethanol and isomers thereof; bisphenol A; 1,8-dihydroxy biphenyl and isomers thereof; and 1,3-bis (2-hydroxyethoxy)benzene.

A polyester useful in the birefringent dielectric multilayer films is polyethylene naphthalate (PEN), which can be made, for example, by reaction of naphthalene dicarboxylic acid with ethylene glycol. Polyethylene 2,6-naphthalate (PEN) is frequently chosen as a first polymer. PEN has a large positive stress optical coefficient, retains birefringence effectively after stretching, and has little or no absorbance within the visible range. PEN also has a large index of refraction in the isotropic state. Its refractive index for polarized incident light of 550 nm wavelength increases when the plane of polarization is parallel to the stretch direction from about 1.64 to as high as about 1.9. Increasing molecular orientation increases the birefringence of PEN. The molecular orientation may be increased by stretching the material to greater stretch ratios and holding other stretching conditions fixed. Other semicrystalline naphthalene dicarboxylic polyesters suitable as first polymers include, for example, polybutylene 2,6-Naphthalate (PBN), polyethylene terephthalate (PET), and copolymers thereof. Non-polyester polymers are also useful in creating polarizer films. For example, polyether imides can be used with polyesters, such as PEN and coPEN, to generate a multilayer reflective mirror. Other polyester/non-polyester combinations, such as polyethylene terephthalate and polyethylene (e.g., those available under the trade designation Engage 8200 from Dow Chemical Corp., Midland, MI), can be used. Suitable first polymers are described, for example, in WO99/36248, WO99/36262; WO 01/22130, and U.S.S.N. 09/444,756.

A preferred first polymer is a coPEN derived with carboxylate subunits derived from 90 mol% dimethyl naphthalene dicarboxylate and 10 mol% dimethyl terephthalate and glycol subunits derived from 100 mol% ethylene glycol subunits and an intrinsic viscosity (IV) of 0.48

dL/g. The index of refraction is approximately 1.63. The polymer is herein referred to as low melt PEN (90/10). Another preferred first polymer is a PET having an inherent viscosity of 0.74 dL/g, available from Eastman Chemical Company (Kingsport, TN).

The other required polymer, referred to herein as the second polymer, should be chosen so that in the finished film, the refractive index, in at least one direction, differs significantly from the index of refraction of the first polymer in the same direction. Because polymeric materials are typically dispersive, that is, the refractive indices vary with wavelength, these conditions must be considered in terms of a particular spectral bandwidth of interest. It will be understood from the foregoing discussion that the choice of a second polymer is dependent not only on the intended application of the multilayer optical film in question, but also on the choice made for the first polymer, as well as processing conditions.

The second optical layers can be made from a variety of second polymers having glass transition temperature compatible with that of the first polymer and having a refractive index similar to the isotropic refractive index of the first polymer. Examples of suitable polymers include vinyl polymers and copolymers made from monomers such as vinyl naphthalenes, styrene, maleic anhydride, acrylates, and methacrylates. Examples of such polymers include polyacrylates, polymethacrylates, such as poly (methyl methacrylate) (PMMA), and isotactic or syndiotactic polystyrene. Other polymers include condensation polymers such as polysulfones, polyamides, polyurethanes, polyamic acids, and polyimides. In addition, the second optical layers can be formed from polymers and copolymers such as polyesters and polycarbonates.

Preferred second polymers are homopolymers of polymethylmethacrylate (PMMA), such as those available from Ineos Acrylics, Inc., Wilmington, DE, under the trade designations CP71 and CP80, or polyethyl methacrylate (PEMA), which has a lower glass transition temperature than PMMA. Additional preferred second polymers include copolymers of PMMA (coPMMA), such as a coPMMA made from 75 wt% methylmethacrylate (MMA) monomers and 25 wt% ethyl acrylate (EA) monomers, (available from Ineos Acrylics, Inc., under the trade designation Perspex CP63), a coPMMA formed with MMA comonomer units and *n*-butyl methacrylate (nBMA) comonomer units, or a blend of PMMA and poly(vinylidene fluoride) (PVDF) such as that available from Solvay Polymers, Inc., Houston, TX under the trade designation Solef 1008. Yet other preferred second polymers include polyolefin copolymers such as poly (ethylene-co-

octene) (PE-PO) available from Dow-Dupont Elastomers under the trade designation Engage 8200, poly (propylene-co-ethylene) (PPPE) available from Fina Oil and Chemical Co., Dallas, TX, under the trade designation Z9470, and a copolymer of atactic polypropylene (aPP) and isotactic polypropylene (iPP) available from Huntsman Chemical Corp., Salt Lake City, UT, under the trade designation Rexflex W111. Second optical layers can also be made from a functionalized polyolefin, such as linear low density polyethylene-g-maleic anhydride (LLDPE-g-MA) such as that available from E.I. duPont de Nemours & Co., Inc., Wilmington, DE, under the trade designation Bynel 4105.

Particularly preferred combinations of first/second polymers for optical layers in IR reflective mirrors include PEN/PMMA, PET/PMMA or PET/coPMMA, PEN/Ecdel, PET/Ecdel, PEN/sPS, PET/sPS, PEN/coPET, PEN/PETG, and PEN/THV. Ecdel is a trade designation for a copolyester ether elastomer available from Eastman Chemical Company (Kingsport, TN). THV is a trade designation for a fluoropolymer commercially available from 3M Company, St. Paul, MN. PMMA refers to polymethyl methacrylate, coPET refers to a copolymer or blend based upon terephthalic acid (as described above), and PETG refers to a copolymer of PET employing a second glycol (usually cyclohexanedimethanol). sPS refers to syndiotactic polystyrene.

For mirror films, a match of the refractive indices of the first polymer and second polymer in the direction normal to the film plane is preferred, because it provides constant reflectance with respect to the angle of incident light (that is, there is no Brewster's angle). For example, at a specific wavelength, the in-plane refractive indices might be 1.76 for biaxially oriented PEN, while the film plane-normal refractive index might fall to 1.49. When PMMA is used as the second polymer in the multilayer construction, its refractive index at the same wavelength, in all three directions, might be 1.495. Another example is the PET/Ecdel system, in which the analogous indices might be 1.66 and 1.51 for PET, while the isotropic index of Ecdel might be 1.52.

The above birefringent dielectric multilayer optical films can be made according to WO99/36248. These films solve the problems of cracking that are typically seen when metalized solar rejection films are nip roll laminated to glazings having compound curvature. The films may wrinkle during nip roll lamination, however, especially when applied to large laminates and/or glazings with severe compound curvatures. In another aspect of the present

invention, the process conditions can be controlled to produce a film with improved shrinkage properties, resulting in a film that does not wrinkle during the nip roll lamination process.

Techniques for manufacturing polymeric multilayer films are described in detail in WO99/36248. Following extrusion and layer formation, the film is cooled on a casting wheel to form a web. The web is then stretched longitudinally (along the web path, or in machine direction, MD) and transversely (across the web path, or in transverse direction, TD) sequentially in a length orienter (LO) and in a tenter. The stretch ratios are determined with reference to the optical and mechanical properties required for a particular application. In a typical LO, the web is preheated between temperature-controlled rollers to a temperature less than the T_g of the first polymer in the film construction. The web is then stretched in a stretch gap under IR lamps that heat the web to a temperature higher than the T_g of the first polymer. The LO oriented film is cooled over cold rollers and is then fed into a tenter.

A tenter typically has four zones, pre-heat, stretch, heatset and cooling zones. Each of these zones may have sub-zones. Length oriented film enters the tenter pre-heat zone and is clipped in chains on each edge of the film. The chains move at the speed of the film. The preheated film moves into the stretch zone where the chains diverge (controlled by rail positions) and is stretched in the transverse direction. The stretched film then passes through the heatset zones to impart additional crystallinity in the film and establish its shrinkage and other mechanical properties before exiting the tenter through a cooling zone. The fully oriented film is finally trimmed and wound on a winder.

If the non-planar substrate to which the film is to be laminated has a specific shape or curvature, to reduce wrinkling the shrinkage of the film may be individually controlled in each in-plane direction. If the curvatures along two principal axes of a region of the substrate are not equal, to laminate the film to that region without wrinkles, the shrinkage of the film may be controlled such that it differs in each in-plane direction. The in-plane direction of the film with the greater shrinkage should be aligned with the dimension of the substrate having the greater curvature.

For example, to achieve substantially wrinkle-free lamination to a non-planar, curved or compound curved substrate, for a PEN or PET based film the shrinkage is greater than about 0.4% in both in-plane directions, preferably greater than about 0.7% in at least one in-plane

direction, and more preferably greater than about 1% in at least one in-plane direction. The shrinkage should be maintained at a minimum to reduce edge delamination. This phenomenon is called a "pull-in." So, the shrinkage is preferably less than about 3% in each in-plane direction, more preferably less than about 2.5% in each in-plane direction.

To introduce shrinkage into the film, for a similar residence time, the tenter heatset temperature is reduced, compared to the heatset temperature for a film intended for lamination to a planar substrate. However, this reduced heatset temperature may result in lower interlayer adhesion. The goal is to find process conditions that balance this trade-off and provide increased shrinkage in combination with acceptable interlayer adhesion.

To minimize shrinkage, it is desirable to select heatset conditions, such as, for example, temperature and/or residence time, that maximize crystal growth rate after the film is oriented. This temperature depends on the materials in the film and is typically between the glass transition temperature and melt temperature of the polymer in the film that has the highest T_g in the construction, typically the first polymer and/or the polymer making up the skin layers, referred to herein as the dominant polymer. As mentioned above, the skin layers can be the same polymer as the first polymer or can be a different polymer. However, the T_g of the skin layers is preferably either equal to or less than the T_g of the first polymer.

For example, typical heatset conditions for PEN based multilayer IR mirror films intended for lamination to a planar substrate are about 249°C. To control shrinkage and provide a film suitable for wrinkle free lamination to a non-planar substrate, the heat set temperature for a PEN based multilayer IR mirror film should be reduced to about 199°C to about 204°C, preferably about 202°C, for about 10 seconds. The heat set temperature for a PET based multilayer IR mirror film suitable for lamination to a non-planar substrate should be about 227°C to 243°C, preferably about 235°C to about 241°C for about 10 seconds.

In addition to the heatset temperature and residence time, another parameter that affects film shrinkage is toe-in. Toe-in is defined as the decrease in rail spacing in the tenter heatset zone measured relative to a maximum rail setting. For example, if maximum width of the film is 170.2 cm at the end of the stretch zone, and film width in the heatset zone is 165.1 cm, then the toe-in is 5.08 cm. The first rail in the heatset zone is preferably set between the maximum rail setting and the rest of the heatset zone rail settings to ensure a smooth transition. However,

tenter rail configuration and tenter width may vary widely, and ideal rail settings must be determined experimentally in each case.

Toe-in affects the shrinkage in both in-plane directions, TD and MD. In addition, there are non-linear interactions between toe-in and heatset temperature for shrinkage properties. The measured shrinkage value is also a strong function of temperature and time used in the measurements. The non-linearity between the measured shrinkage values and heatset temperature and toe-in can be very different for different measurement conditions. In the present application, shrinkage properties are measured using a standard test condition of 150°C for 15 minutes.

Again, the toe-in required depends on the materials in the film, on the intended application and on the heatset temperatures and residence times. Either one or the other or both heat set temperature and the toe-in may be varied to control the shrinkage in the film. For example, for a PEN based multilayer IR mirror film, a statistical model predicts that to get MD shrinkage of about 1% and TD shrinkage of about 2%, the tenter heat set temperature should be about 202°C for about 10 seconds and the toe-in should be about 1.27 cm to about 2.54 cm from a maximum rail setting of about 177.8 cm.

To control shrinkage and provide a film suitable for wrinkle free lamination to a non-planar substrate, the toe-in should be adjusted depending on the polymer used. For a PEN based multilayer IR mirror film, the toe-in should be about 0 to 3.81 cm, preferably about 1.27 to 2.54 cm at the preferred heatset temperature, film width and heatset zone residence times mentioned above. For a PET based multilayer IR mirror film, the toe-in should be about 0 to 5.08 cm, preferably about 0 to 3.81 cm at the preferred conditions described above.

The multilayer IR film may be coated with a primer to promote adhesion to the PVB. Coatings may change the shrinkage properties of a film. Typically, a primer coating may reduce TD shrinkage by about 0.2% to about 0.4% and increase MD shrinkage by about 0.1 to about 0.3%. The amount of change depends on the drying temperature of the coater oven, as well as on film tension and residence time in the heatset zone.